Radioiodine Speciation and Immobilization at the Savannah River Site, Hanford Site, and Fukushima Prefecture

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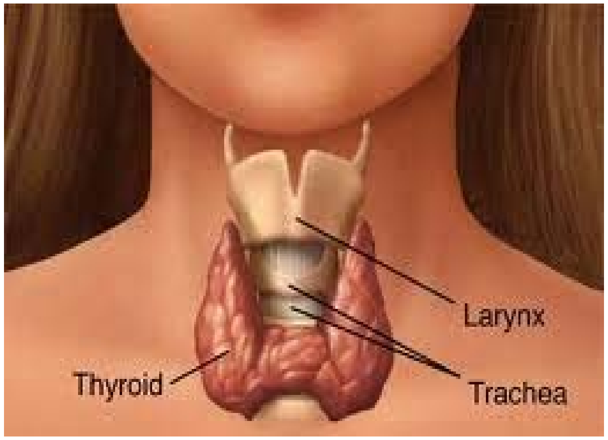
Radioiodine: Importance of Speciation

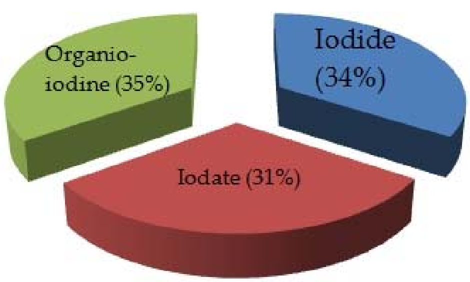
- I^-, IO_3^-, and organo-I commonly exists together in same system
- We're the only lab in the DOE system that can do iodine speciation at ambient concentration: at MCL concentration
- Biogeochemistry:
  - *Soil sorption (Kd): I^- = 0.5, IO_3^- = 5 and organo-I = 50 mL/g.*
  - *Precipitation: $I^-_{(aq)} + Ag_{(aq)} \rightarrow AgI_{(s)}$ (but not IO_3^-),*
  - *Co-precipitation $IO^-_{3\,(aq)} + CaCO_{3(aq)} \rightarrow Ca(IO_3)CO_{3(s)}$*
- **Risk: ^{129}I at SRS accounted for only 0.00002% of the radioactivity released from the site, yet it comprised 13% of the offsite population dose**
  - *14-g thyroid contains 90% of body's iodine*
  - ***MCL = 0.04 Bq/L; among the lowest of all rads***
  - *#1 cause of cancer from Chernobyl accident , ^{131}I*
  - **Risk: ^{129}I >>>> $^{129}IO_3^-$ > org-I**

Program Objective: Iodine speciation of a system needs to be taken into consideration before engineering remediation and waste disposal solutions.

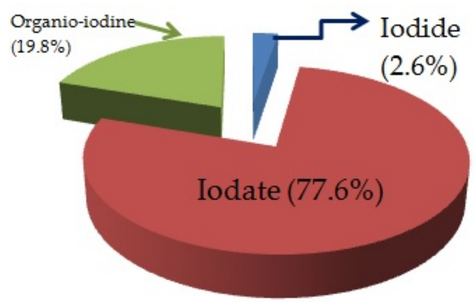


Iodine Speciation

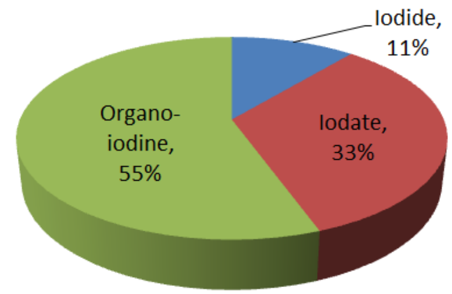
Savannah River Site	Hanford Site	Fukushima Prefecture



Otosaka et al. 2011
(JAEA)



Zhang et al. 2014

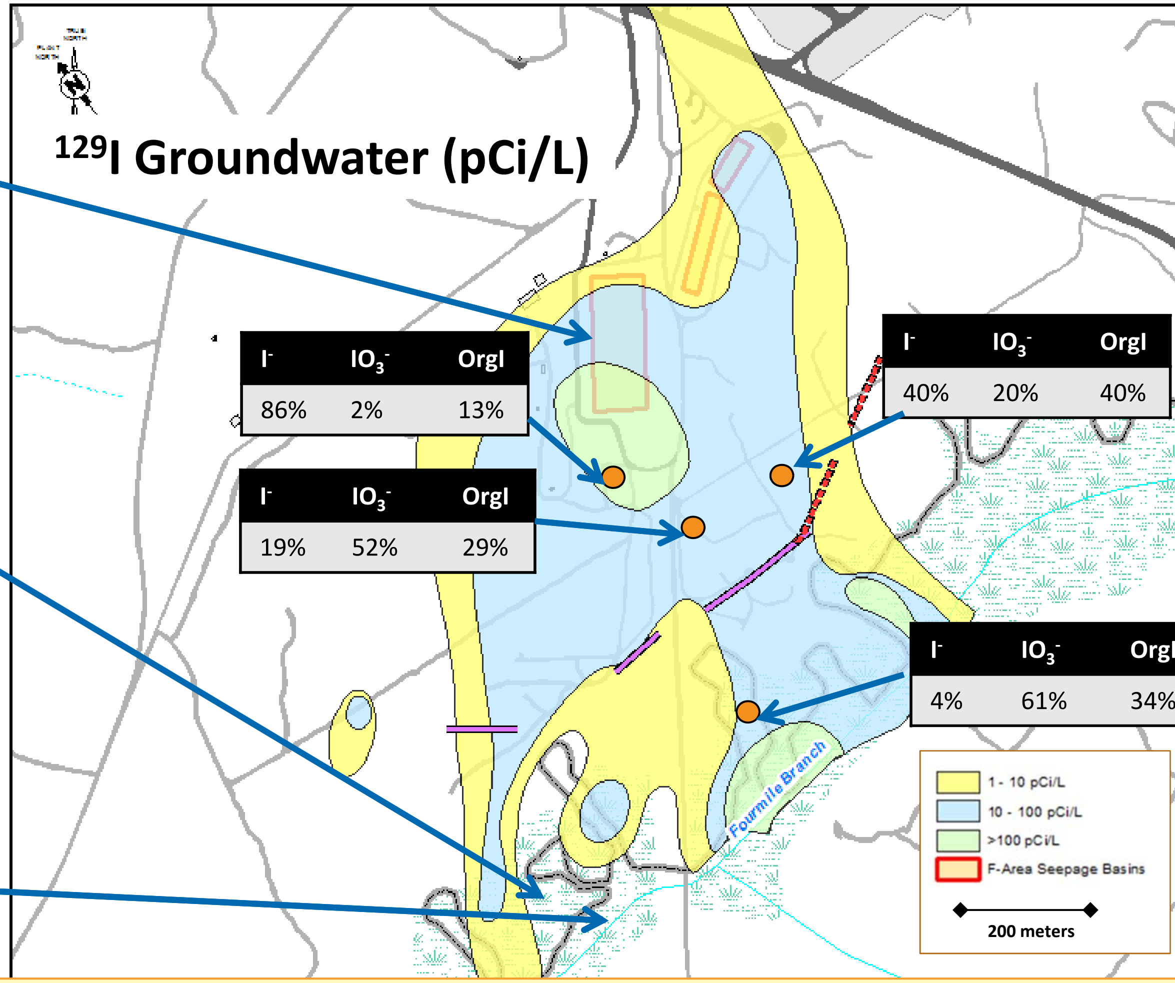


Xu et al. 2014
(Kyoto, Hyogo, Kobe Universities)

- ❑ ***Multiple iodine species exists in the same sample***
- ❑ ***Iodine speciation varies based on local geochemistry.***
- ❑ ***Natural organic matter control iodine sorption***

F-Area Field Radionuclide Testbed

Contaminated with ^{3}H, ^{137}Cs ^{129}I, ^{99}Tc, U, Am, Cm, Pu, NO_3^-



I^-	IO_3^-	OrgI
86%	2%	13%

I^-	IO_3^-	OrgI
19%	52%	29%

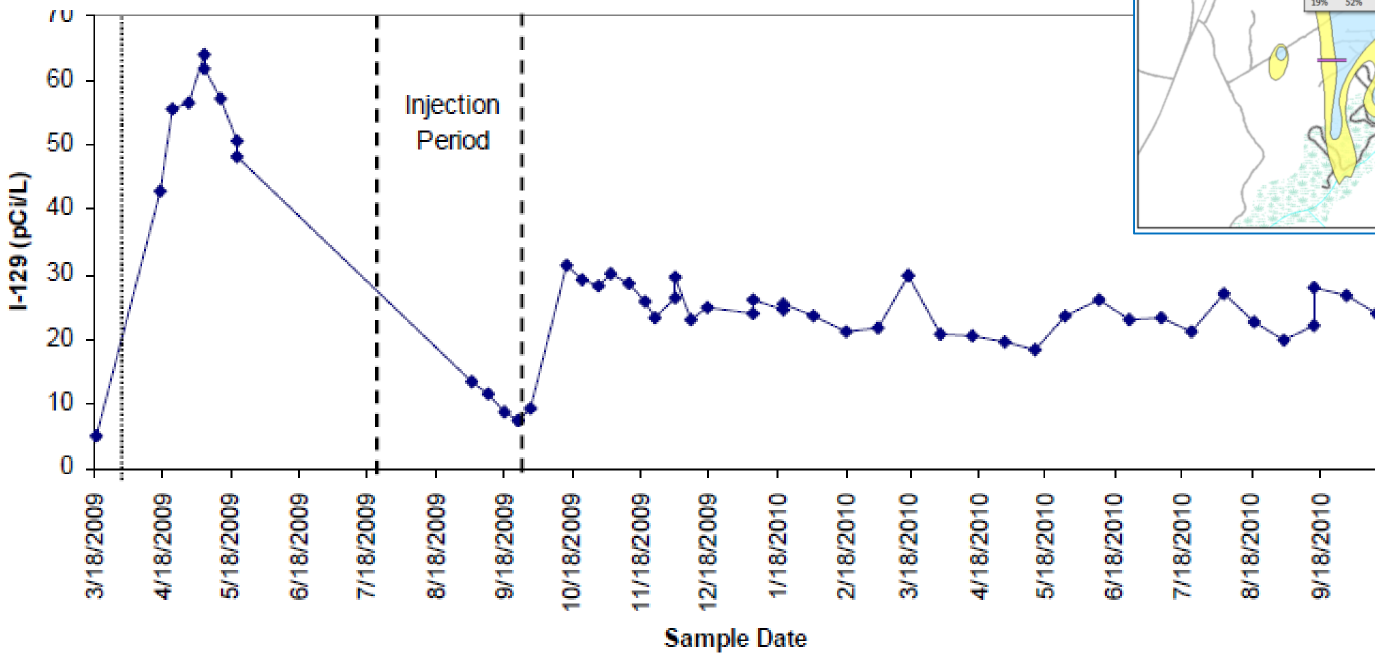
I^-	IO_3^-	OrgI
40%	20%	40%

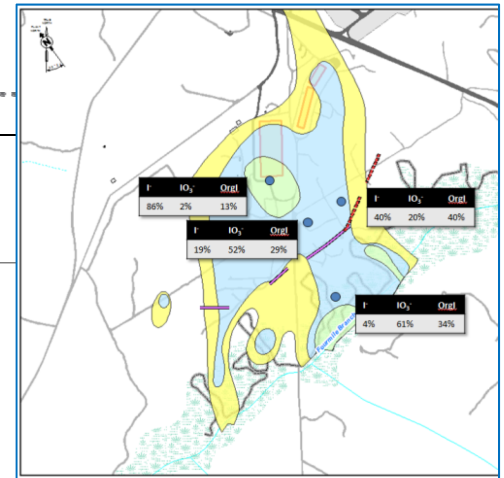
I^-	IO_3^-	OrgI
4%	61%	34%

- **There is a highly mobile fraction of iodine that everyone is aware of based on groundwater monitoring results.**
- **However, there is a second and third fraction, that is less mobile that is not presently accounted for.**

Example of ^{129}I Field Demonstration

AgCl injection into ^{129}I plume in F-Area

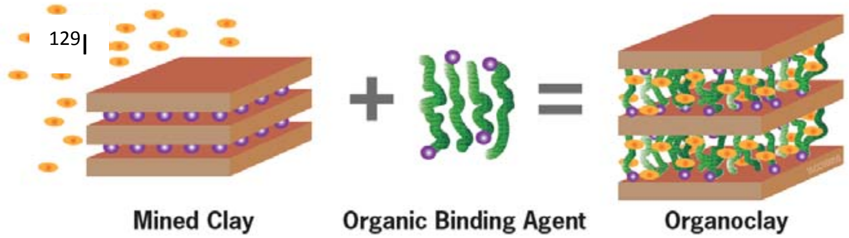




$$I^- + AgCl_{(aq)} \rightarrow AgI_{(s)}$$

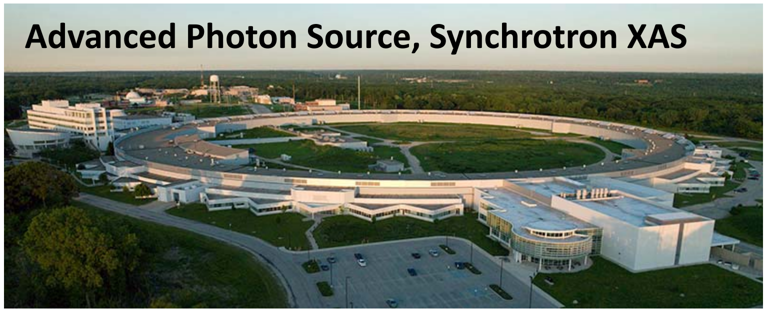
Demonstrated *in situ* technology by forming the sparingly soluble AgI precipitate.

Iodine – Highly Effective, Low-cost Sorbents



- Sorption (Kd = $I_{solid}/I_{groundwater}$):
  - Organo-Clay: Kd >20,000 mL/g $9/kg
  - GAC: Kd >10,000 mL/g $25/kg
  - P-Biochar: Kd = 55,000 mL/g $9/kg
- Kinetic: Fast reaction
- Adsorption capacity: >3.2 mg/g for I removal from wide pH range

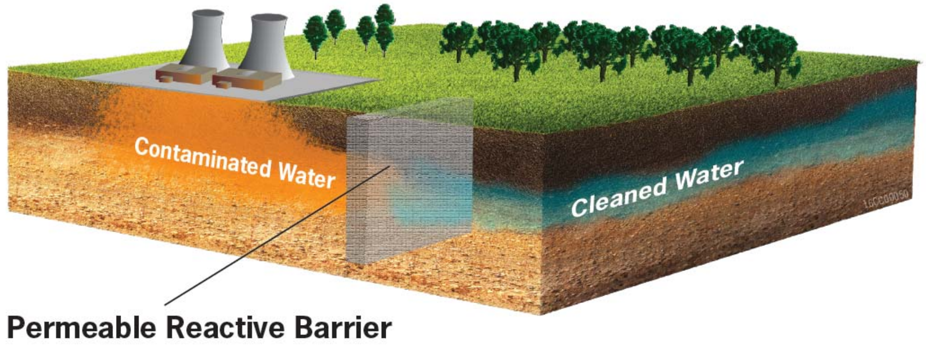
Advanced Photon Source, Synchrotron XAS

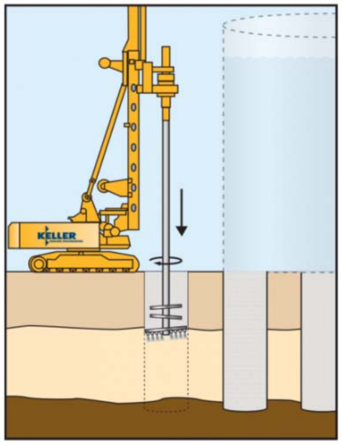


Key Takeaways

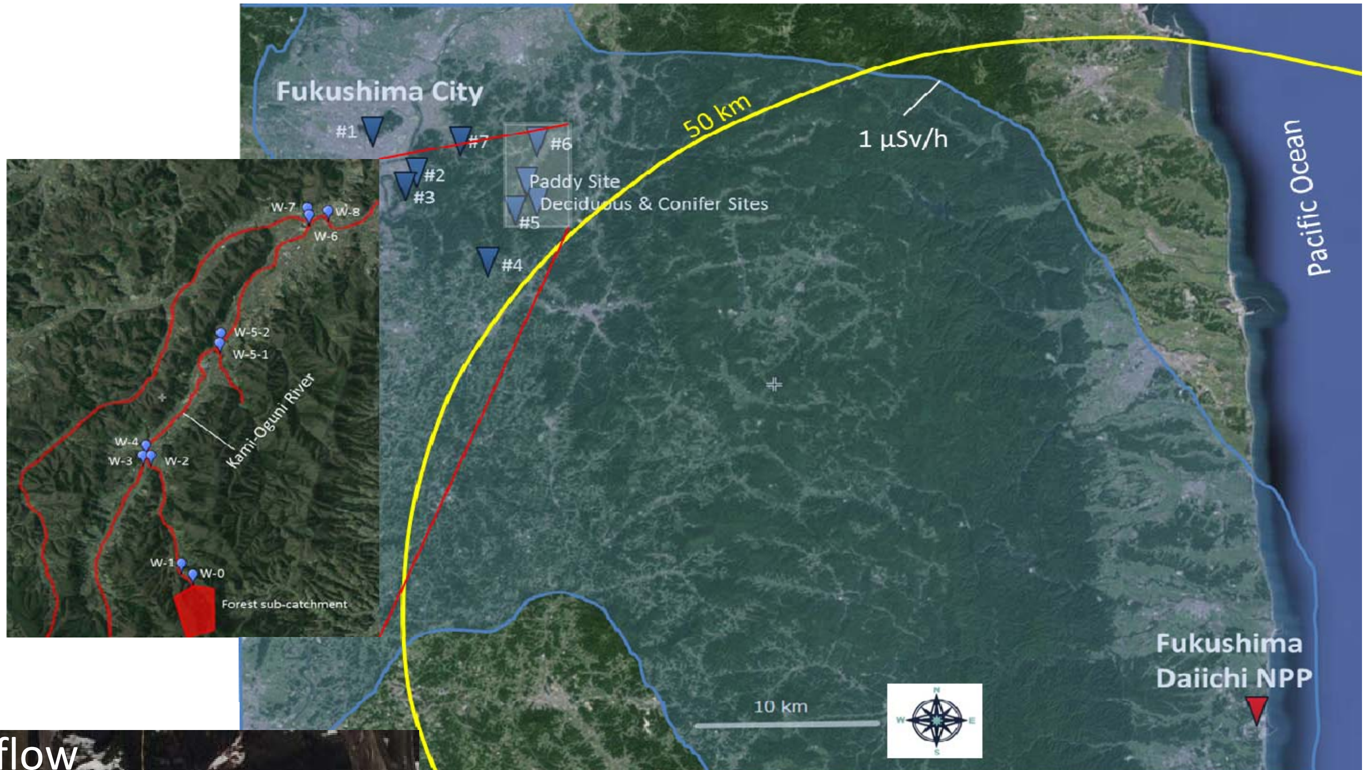
- Iodine species sorbed: Iodine speciation influences the degree that sorbents are effective

Motivation:



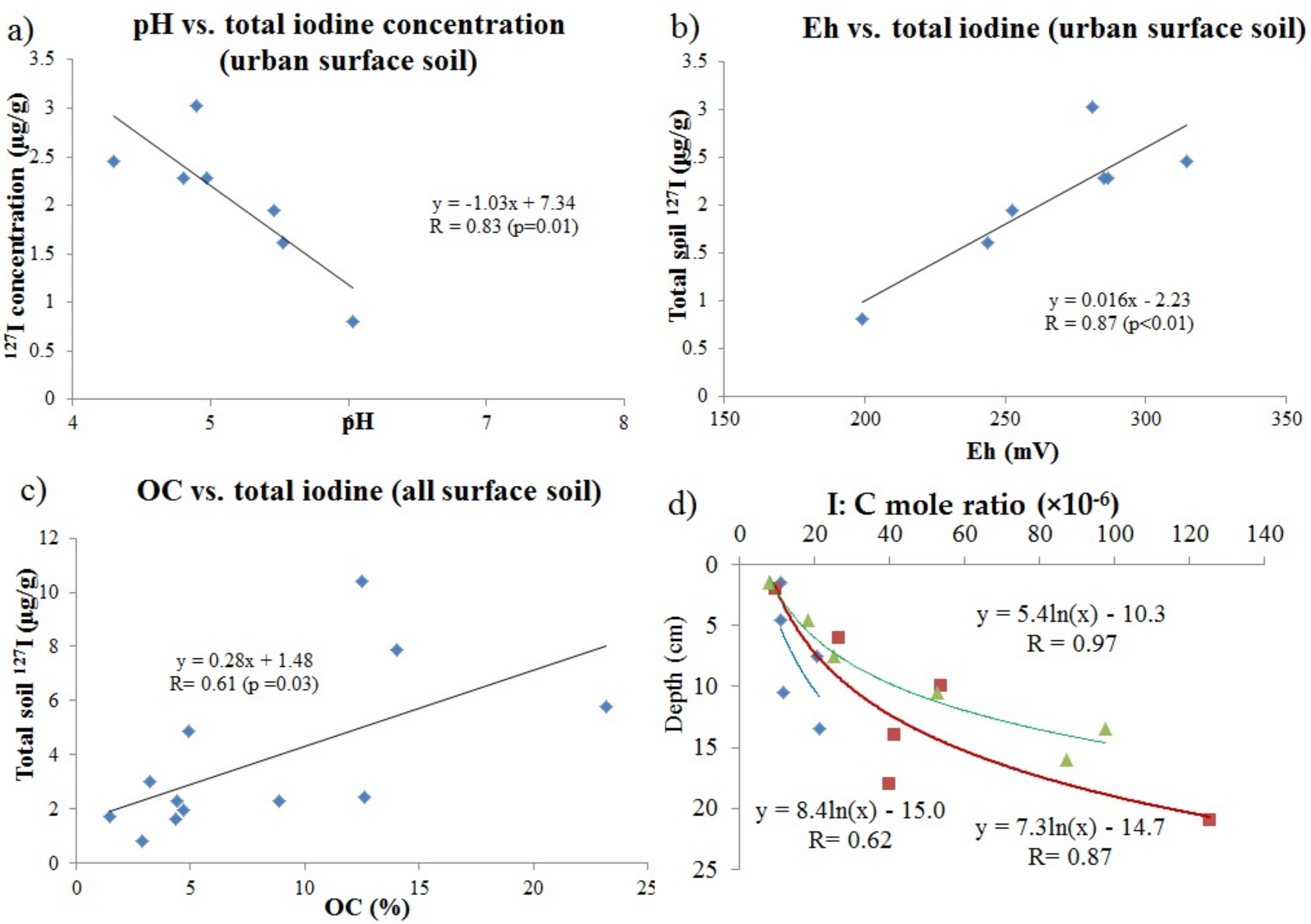


Iodine Speciation Transformations: Precipitation → Throughfall → Stemflow → Soil



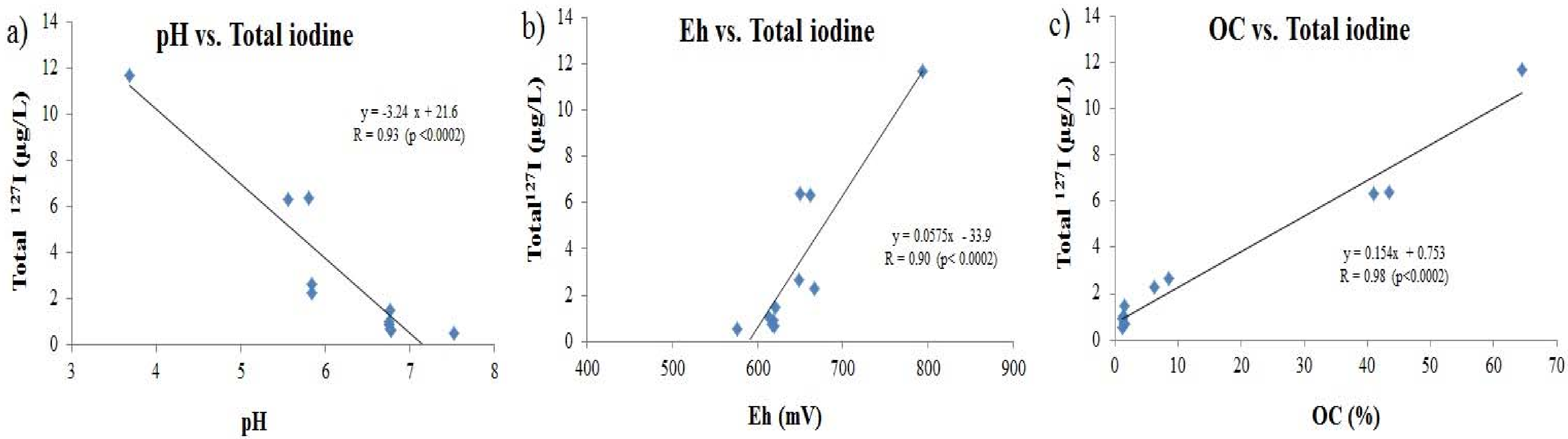
- <u>Soil samples</u>: 7 urban area; 1 paddy field, 1 deciduous forest, and 1 coniferous forest.
- <u>Surface water samples: A</u>long the flow path from a tributary of the Oguni River
- Precipitation → stemflow → throughfall

Factors affecting I conc. and speciation in soil



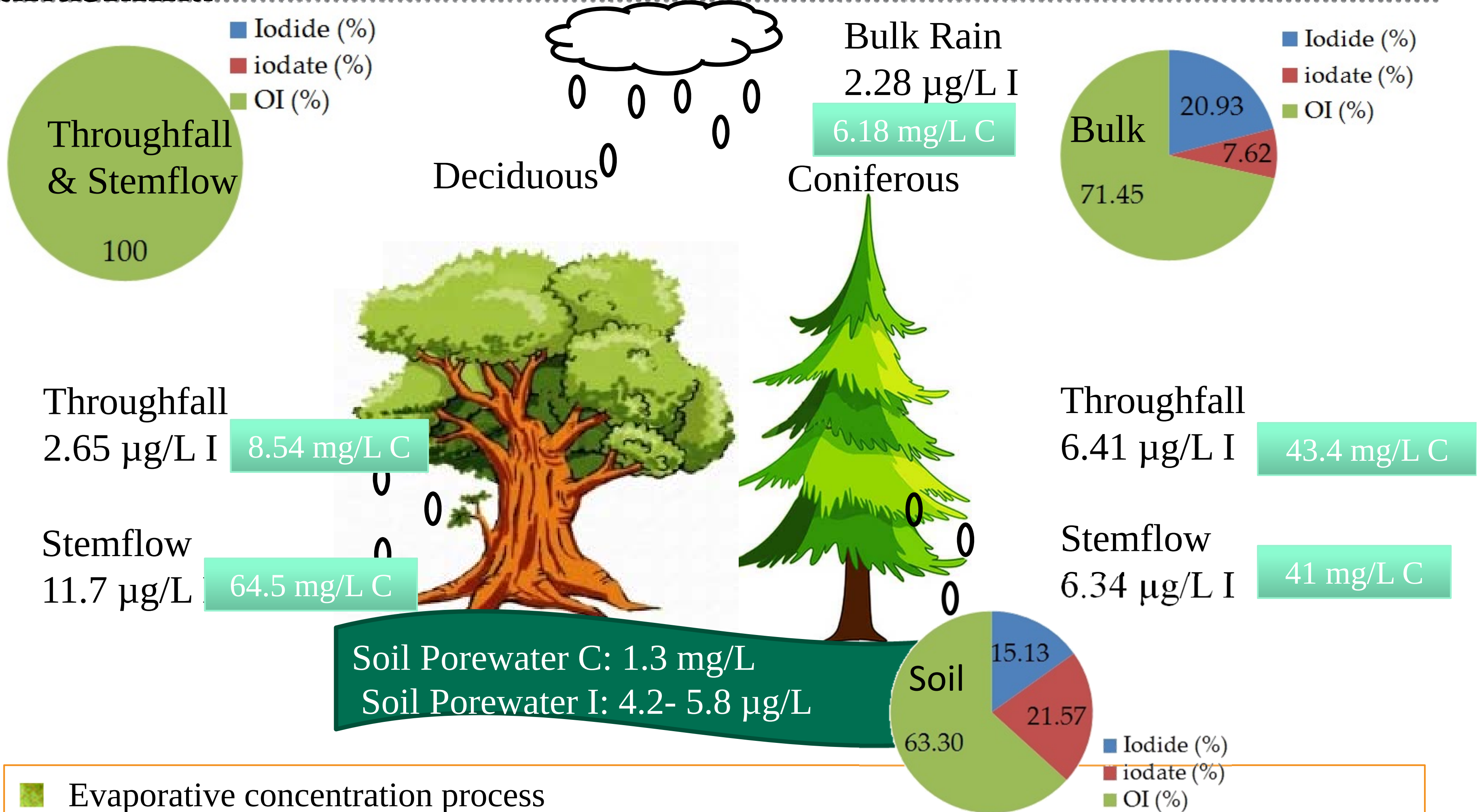
- Soil total I conc. is controlled by pH, Eh, organic matter and their composition.

Factors affecting I conc. and speciation in rainwater



pH, Eh and OC cooperatively affect the geochemical behavior of iodine in surface water and wet deposition.

Iodine concentration, speciation and risk transformations near Fukushima



- Evaporative concentration process
- Scavenging fine mists which are enriched in iodine yet might not be collected by the open rainfall
- Increase in OC contents and enrichment in aromatics and refractory condensed aromatics that fix iodine—formation of organo-iodine

Summary

- Environmental and disposal decisions of radioiodine must be based on iodine speciation and not just total iodine concentrations.
- Iodine speciation greatly impacts the mobility and risk posed by radioiodine.
- Japanese – U.S. collaborations have resulted in significant advances in the environmental understanding and the immobilization of radioiodine (4 manuscripts/2 projects/1 undecided proposal/>10 presentations).